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DEGENERATE FOUR-WAVE MIXING
LINESHAPES IN SODIUM VAPOR UNDER PULSED EXCITATION

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ABSTRACT

Degenerate four-wave mixing (DFWM) lineshapes are investigated in sodium vapor near the D_2 resonance line using nearly Fourier transform limited pulses. At low pump intensities sub-Doppler resolution is obtained. When the Rabi frequency associated with the pump intensity becomes equal to the ground state hyperfine frequency separation of sodium, each component of the double-peaked DFWM spectrum further splits into two components each. Adiabatic following model explains the near-resonant intensity dependence of the DFWM signal but is insufficient to explain the whole structure.

Resonant degenerate four-wave mixing (DFWM) has been shown to be an efficient technique for the generation of phase conjugate wave fronts[1]. In backward configurations, where the two pump beams as well as the two signal (probe and conjugate) beams counter-propagate to each other, the DFWM spectrum is Doppler free and has been used for high resolution spectroscopy[2]. However, true spectral information is obtained only when the pump beam intensities are small compared to the saturation intensity of the resonant medium. At higher pump intensities, complicated line shapes are observed; in particular, the line broadens and splits into two components. Several experimental and theoretical studies of this behavior have appeared in recent literature[3]. Most of the studies have employed cw lasers and hence steady state analyses have been used to explain the observed behavior.

The early experiments on resonant DFWM employed pulsed dye lasers[4]. DFWM spectrum in this case is also complicated and shows a dip to zero at the line center. Bloom et al. were the first to observe such a line shape. In the pulsed case, fluctuations (pulse to pulse energy, pulse-shape, and center frequency fluctuations) and other nonlinear effects such as self focussing complicate the line shape even further. Recently we have measured the DFWM line shapes with a pulsed dye laser system which was stabilized in center frequency and bandwidth. Our control of the fluctuations in the DFWM signal was essential for the measurement of the photon counting statistics of light generated via DFWM[5]. In this letter we describe the results of a study of DFWM line shapes with such a source.

The experimental setup is essentially the same as that in Ref. 6 and is shown in Fig. 1. An externally stabilized cw ring dye laser (sub MHz line width) is amplified through a chain of pulsed dye laser amplifiers pumped by the smoothed output of a Nd:YAG laser[7]. The output pulses of 4 ns duration with typically 10-20% energy fluctuations and a total line width of 200 MHz which is approximately twice the Fourier transform limited line width are used to perform DFWM.

About 10-50 MW/cm^2 pulse intensities are available from such a system. Backward DFWM geometry is employed with orthogonally polarized pump beams[8]. Phase conjugate (PC) signal whose polarization is orthogonal to the probe beam (PB) is separated using a polarization beam splitter (PBS) and directed onto a photomultiplier (PMT) whose output is sent to a boxcar integrator. The output of the boxcar which is proportional to the PC pulse energy goes to the y-axis of a chart recorder whose x-axis is swept with the dye laser frequency. The boxcar whose gate duration is chosen to be 80 ns is triggered optically. The PC signal is first delayed optically and then electrically using a 50 Ω cable inserted between the PMT and the boxcar in such a way that it arrives in the middle of the gate duration.

DFWM is performed in sodium vapor generated in a heat-pipe oven. It is maintained at 310°C implying a sodium vapor density of 4×10^{14} atoms/cm^3 for the measurements reported in this letter. 2.2 Torr of helium is used as the buffer gas. All the beams arrive in time coincidence at the sodium cell. The dye laser frequency is scanned over 10 GHz across the sodium D_2 line. The spectra obtained in this way are shown in Fig. 2 as a function of the pump beams intensities which are kept the same (to within 10%) for both the pumps. Appropriate neutral density filters are introduced in the probe beam path to avoid saturation of the PMT. The probe pulse energy is kept less than a few percent of the pump pulse energy in all cases to avoid pump depletion.

At the lowest pump intensities used of 0.3 kW/cm^2 (all relative intensity measurements are ± 10%)[9], the line shape consists of two peaks which are 4.5 GHz apart as shown in Fig. 2a. Also shown is the fluorescence observed in a direction making a small angle ($\simeq 1°$) with the PC beam. The pulse widths (FWHM) are 0.74 and 0.83 GHz for the lower and the higher frequency peaks respectively. Therefore sub-Doppler resolution is obtained at these pump intensities even though the peaks occur away from the center of the Doppler-broadened line of width 1.8 GHz.

As the pump intensities are increased to 0.6 kW/cm^2, the higher frequency peak splits into two components separated by 1.3 GHz as shown in Fig. 2b. A further increase of the pump intensities to 1.9 kW/cm^2 leads to a splitting in the lower frequency peak as well, as shown in Figs. 2c and 2d. In Fig. 2d the reflectivity at the highest frequency peak is 0.7%. The reason for such a low reflectivity is because a relatively large angle was chosen between the pumps and the probe beam[2]. This choice was dictated by the low background requirement in our DFWM quantum noise measurements reported earlier[5]. With smaller angle and higher pump intensities (at least an order of magnitude higher than those reported herein) we have observed reflectivities as large as 700. Under these conditions the conjugate pulse duration is significantly shorter than the probe pulse duration. A systematic study of this behavior has recently been reported[10].

At still higher pump intensities of 3-10 kW/cm^2, only two peaks remain as shown in Figs. 2e and 2f. A further increase of the pump intensities leads to a broadening and weakening of the lower frequency peak which is consistent with the observations of earlier workers. A slight broadening and weakening is already observable in Fig. 7g. Jabr et al.[4] did not observe the lower frequency peak with the same choice of the pump and probe beam polarizations that we employ. Their pump intensities were at least an order of magnitude larger than ours. At high pump intensities, self-defocussing of the pump beams on the lower frequency side of the resonance causes a reduction of the actual pump intensities in the mixing medium, which results in a lowering of the PC signal.

The splitting of the DFWM lineshape (Figs. 2b-2d) was not observed in earlier experiments[4]. This we believe is because we have used stable center frequency nearly Fourier transform limited pulses for the above measurements[6]. Furthermore, at low intensities sub-Doppler lines whose widths are limited by power broadening are observed.

The nonlinearity responsible for DFWM in sodium vapor is due to resonantly enhanced electronic Kerr effect. Grischkowsky et al.[4] used the adiabatic following (AF) model for a two level atom under pulsed excitation to derive an expression for the DFWM reflectivity as a function of the pump intensities and detuning. When the AF conditions are satisfied,[11] the third order nonlinear susceptibility is given by

$$\chi^{(3)} = -\frac{N\mu^4A^2}{h^3(\nu-\nu_0)^3\ (1+A^2/A_s^2)^{3/2}} \tag{1}$$

where μ is the electric dipole moment of the two level atom, A is the pump fields amplitudes (assumed equal for both the pumps), $\nu-\nu_0$ is the detuning of the DFWM frequency from the atomic line center frequency, N is the total number of atoms per unit volume, and A_s^2 is the normalized saturation intensity given by $A_s = h|(\nu-\nu_0)|/\mu$. The DFWM reflectivity is given by $R = \tan^2 \kappa L \simeq \kappa^2 L^2$ where $\kappa = 2\pi\nu\chi^{(3)}/2cn$, n is the linear refractive index of the medium, L is the interaction length, and the approximation is valid under conditions of weak reflectivity.

As the pump intensities are increased, the DFWM signal saturates because of the saturation of $\chi^{(3)}$. This is verified for a detuning of 2.3 GHz where the AF conditions are satisfied as shown in Fig. 3. DFWM reflectivities were measured from Fig. 2 at a detuning marked by the arrows. Dots are the experimental data and the solid line is a fit to Eq. (1) for I_s = 7.8 kW/cm^2 which agrees well with $I_s = \epsilon_0 c(h(\nu-\nu_0)/\mu)^2/2$ = 2.1 kW/cm^2. Thus a good agreement is obtained with the AF model in its region of validity.

The detailed lineshapes cannot be predicted using the AF model as it is not valid sufficiently close to resonance. Steady state models[3] developed to explain cw DFWM lineshapes cannot be applied in the transient case of our experiment.

The splitting of the DFWM lineshape as shown in Figs. 2b-2d occurs when the Rabi frequency corresponding to the pump intensities is close to the ground state hyperfine splitting of the sodium atom which is 1.77 GHz. Moreover, the splitting of the lower frequency peak occurs at approximately twice the pump intensities than that of the higher frequency peak. This suggests that the multiple level nature of the sodium atom is playing a role. This is not surprising because the spectrum of the pulses used in this experiment is much narrower than the ground state hyperfine splitting of sodium. For our 200 MHz, 4 ns pulses, sodium can be well modelled as a three level atom of Λ type. The DFWM mechanism in our experiment is more complicated than it seems because at low pump intensities of Fig. 2a, although sub-Doppler resolution is obtained, the peaks are not separated by 1.77 GHz as would be the case in a cw DFWM experiment when the spectrum of the laser used is much narrower than the ground state hyperfine splitting. A detailed comparison with the theory can only be made when Maxwell-Bloch equations are solved in the transient regime for the backward DFWM configuration taking the three level nature of the medium into account.

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9. All intensity measurements refer to peak intensities of the pulses which are spatially in TEM_{00} mode. The absolute value could be off by as much as a factor of 5.

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11. D. Grischkowsky, in Physics of Quantum Electronics, Vol. II, eds. S.F. Jacobs, M. Sargent III, J.F. Scott and M.O. Scully (Addison-Wesley, Reading, 1975) p. 437-452. For nearly Fourier transform limited pulses, the AF condition is

$$\left| A^{-1} dA/dt + T_2^{-1}\left[1 + \{\mu A/h(\nu-\nu_o)\}^2\right]\right| \ll |\nu-\nu_o| \left[1 + \{\mu A/h(\nu-\nu_o)\}^2\right],$$

which is satisfied for 4 ns pulses with $T_2^{-1} = 10$ MHz and $|\nu-\nu_o| = 2.3$ GHz.

Figure Captions:

1. Schematic of the experimental apparatus. YAG = frequency doubled Nd:YAG laser, DL = cw dye laser, M = total reflector, BS = beam splitter, DET = detector, HWP = half wave plate, PBS = polarization beam splitter, PMT = photo-multiplier tube, PC = phase conjugate beam, BP = backward pump beam, FP = forward pump beam, PB = probe beam.

2. DFWM lineshapes at various pump intensities. Vertical scale is arbitrary and is linearly proportional to the DFWM signal, plotter scale factors are labelled. Also superimposed are the fluorescence spectra in parts a, b, and g. Pump intensities are labelled in kW/cm^2 in each plot. Start frequency in parts f and g is slightly shifted.

3. Dependence of the DFWM signal on the pump intensities.

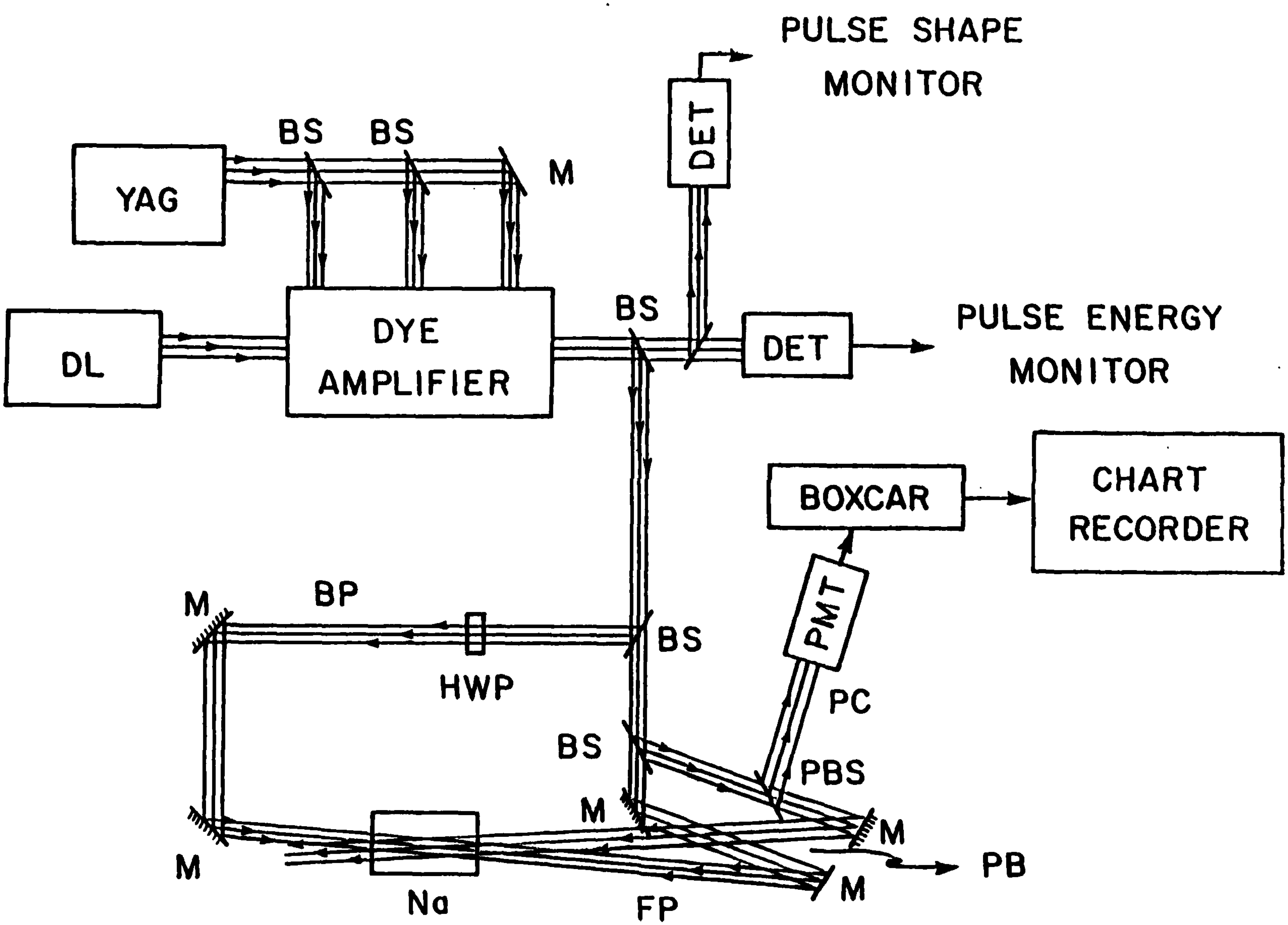
PULSE SHAPE MONITOR
DET
BS
BS
M
YAG
DL
DYE AMPLIFIER
BS
DET
PULSE ENERGY MONITOR
BOXCAR
CHART RECORDER
PMT
M
BP
BS
HWP
PC
BS
PBS
M
M
M
PB
M
Na
FP

Fig. 1

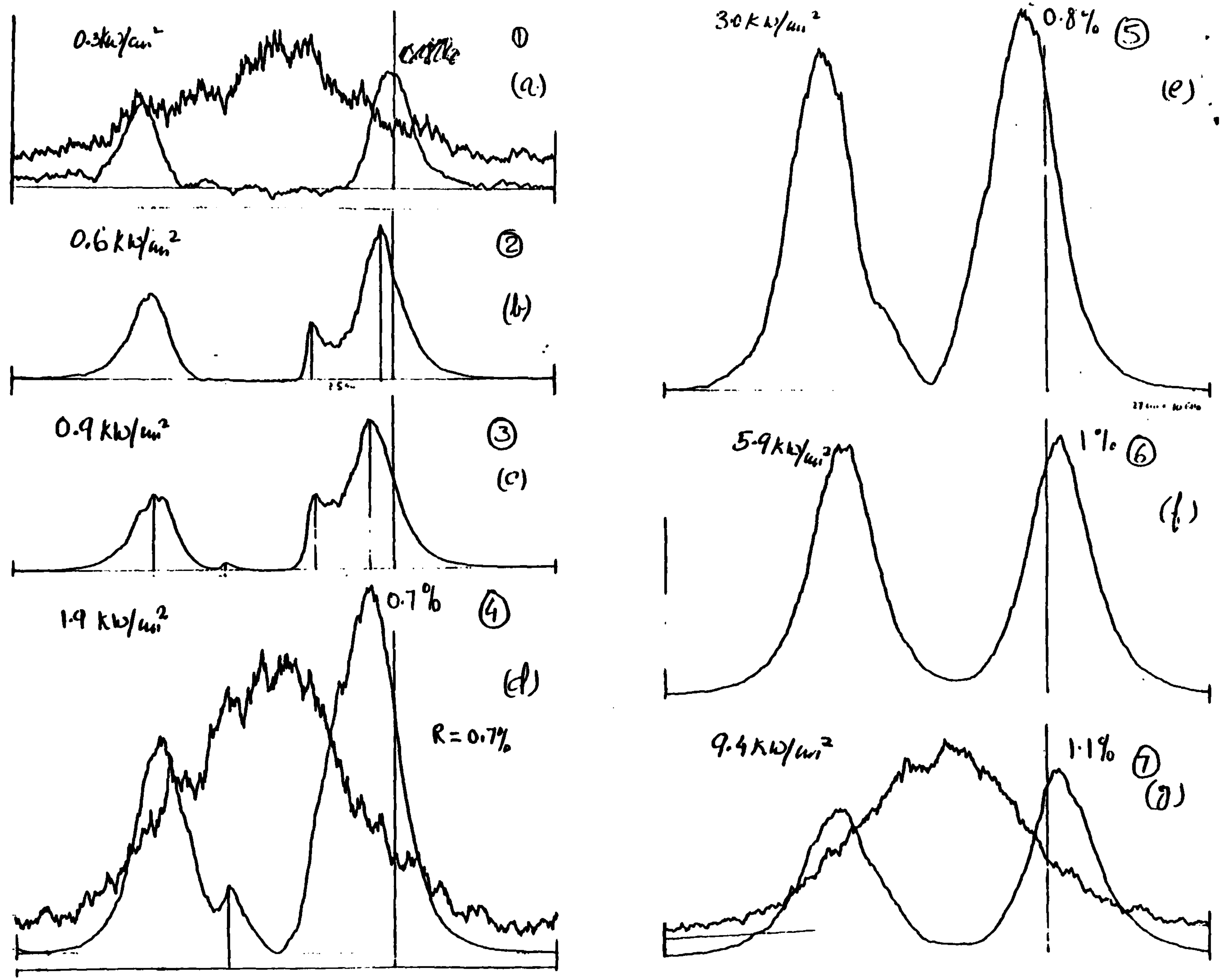
0.3 kW/cm²
① (a)
0.6 kW/cm²
② (b)
0.9 kW/cm²
③ (c)
1.9 kW/cm²
0.7%
④ (d)
R = 0.7%
3.0 kW/cm²
0.8%
⑤ (e)
5.9 kW/cm²
1%
⑥ (f)
9.4 kW/cm²
1.1%
⑦ (g)

Fig. 2

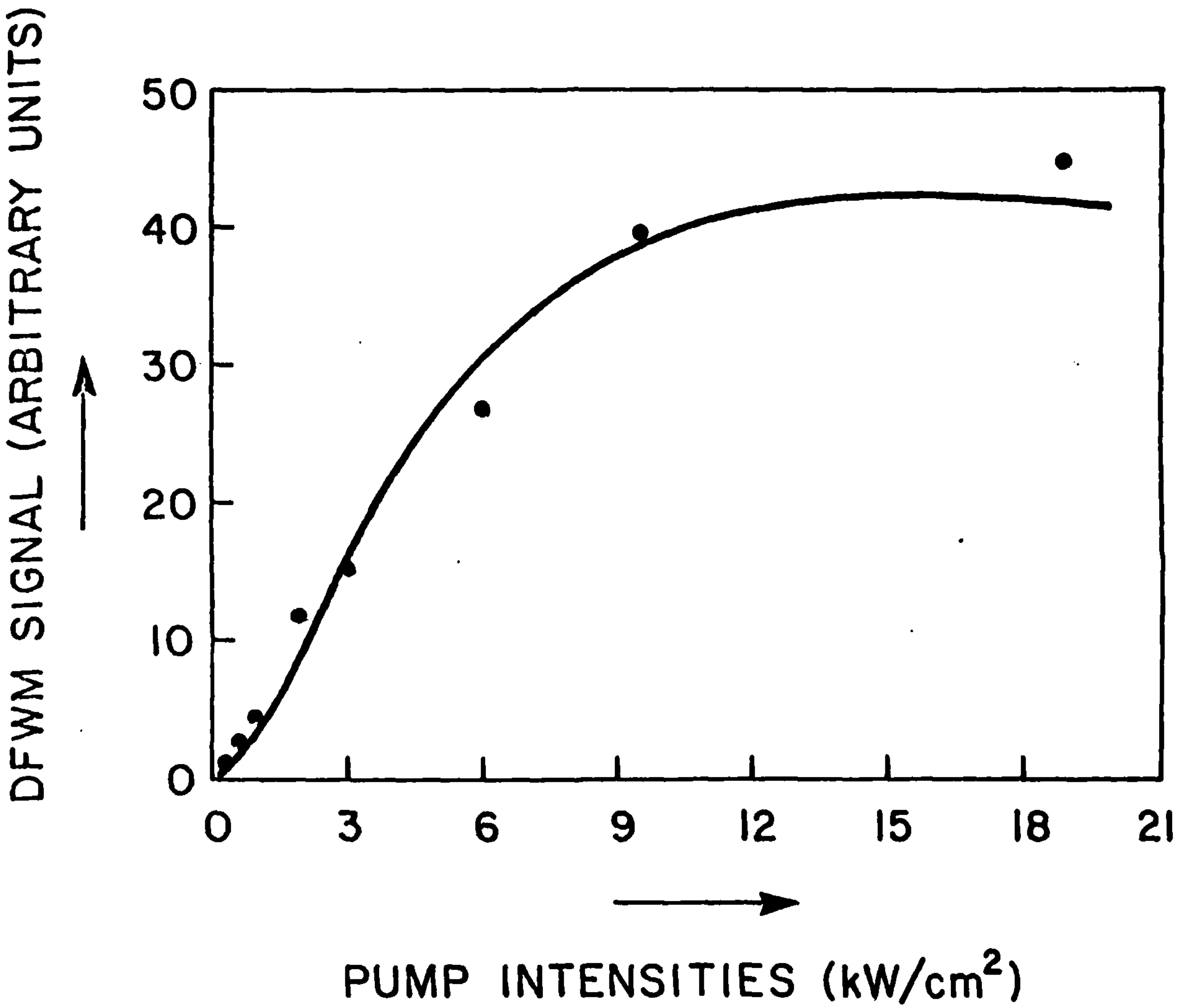
DFWM SIGNAL (ARBITRARY UNITS)
50
40
30
20
10
0
0
3
6
9
12
15
18
21
PUMP INTENSITIES (kW/cm^2)

Fig. 3